Complete theory of the optical shifts of excitons in 2D semiconductors

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Increasing the speed limits of conventional electronics requires innovative approaches to manipulate quantum properties of electrons beyond their charge. A promising alternative is lifting the valley degeneracy of exciton states in transition metal dichalcogenide (TMD) monolayers through coherent optical interactions on timescales corresponding to a few tens of femtoseconds. Specifically, the observed optical Stark and Bloch-Siegert effects (valley-selective blue shifts of excitons by more than 30 meV) generated by strong nonresonant circularly polarized light pave the way for the realistic manipulation of valleys in these materials [1, 2]. These results open the door for ultrafast valleytronics operating at multiterahertz frequencies.

Practical applications of such effects require a comprehensive theory of energy shifts in intravalley excitons generated by strong non-resonant optical fields. Previous theoretical studies of this effect have been incomplete because only the so-called interband coupling term was considered. The validity of this approximation had not been proven earlier, and therefore, the theory of the optical shifts remains incomplete.

We reconsider the theory of optical shifts in 2D semiconductors using the approach of Semiconductor Bloch equations (SBE). We introduce the previously neglected intraband term, which is responsible for electron dynamics within the separate bands, into the SBE. We develop a new perturbation technique and calculate the optical shifts of intravalley exciton energies in TMD monolayers, which can be measured using the pump-and-probe technique [3]. We demonstrate that the intraband term generated by the electric field of the pump pulse provides a nonzero contribution to the optical shift, in addition to the interband contribution. This allows us to analyze the limits of applicability of previous findings. The possibility of experimental measurement of the obtained results is also discussed.

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References

[1] A. O. Slobodeniuk, P. Koutenský, M. Bartoš, F. Trojánek, P. Malý, T. Novotný, and M. Kozák, npj 2D Mater. Appl. **7**, 17 (2023).
[2] A. O. Slobodeniuk, P. Koutenský, M. Bartoš, F. Trojánek, P. Malý, T. Novotný, and M. Kozák, Phys. Rev. B **106**, 235304 (2022).
[3] A. O. Slobodeniuk, T. Novotný, arXiv:2501.06885

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